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ATOMIC PROCESSES RELEVANT TO
ANTIMATTER FUEL
PRODUCTION AND STORAGE

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FINAL REPORT

31 MAY 1994

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<p>Electric field effects have been identified in the dissociative recombination of H_3^+ ions. This process proceeds via the formation of high lying Rydberg states that are field ionized in the ion analyser in our apparatus. Reducing this field results in measured cross section five times larger than with the normal operating field. A brief update on measurements of stimulated radiative recombination of He^+ is presented.</p>					
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CHAPTER 1

OVERVIEW OF PROGRAM

A. INTRODUCTION

The aim of the current research program, as initially proposed, was to study the recombination rates for hydrogen cluster ions and to examine methods by which the recombination rate could possibly be altered. The impetus for this work was to examine the chemistry involved in the development of cluster species such as H_5^+ , H_7^+ etc. from smaller hydrogen molecules such as H_2^+ and H_3^+ . Such clusters would be formed in any scheme developed to produce anti-hydrogen fuel. Since recombination acts as a very effective destruction mechanism for clusters, it is crucial to understand the characteristic of this process so that ways can be found to minimize its effect.

In previous phases of this project^{1,2,3,4}, the laser stimulated radiative recombination of electrons and protons (and of electrons and He^+ ions) was demonstrated. Using the same experimental arrangement, it was shown⁴ that the presence of laser light in the recombination region could lead to the diminution of the recombination cross section for H_3^+ molecules. The reason for this is that the recombination of H_3^+ proceeds through the formation of excited Rydberg states of the neutral H_3 molecule and that these states can be destroyed by photoionization. In a separately funded US Air Force program, the author performed an in depth study of the recombination of H_2^+ and H_3^+ molecular ions⁶. In this study it was found that the cross sections for ground vibrational state H_3^+ ions was

about an order of magnitude lower than that found when the ions were vibrationally excited. This finding contradicts however, recent results from afterglow experiments (see below) and it was surmised that this finding could be due to the destruction of the intermediate molecular states, involved in the recombination, by field ionization effects in the merged beams apparatus. This effect has been studied in greater detail during the past year and the results are reported in this document.

Continued funding for the current project was cancelled due to budgetary difficulties at AFOSR and so the program, as originally proposed, has had to be curtailed. Investigation of the recombination of the cluster species requires the construction of a new, high pressure, ion source on an as yet untested 100 keV accelerator. Given the cancellation of financial support, it was decided not to proceed with this venture and instead to concentrate on (a) investigating field effects in the recombination of H_3^+ and (b) continuing with our investigation of stimulated recombination of He^+ . During the year, a considerable effort went into the investigation of the recombination of HeH^+ ions and it was shown that previous measurement made by us⁷ were in fact attributable to the recombination of metastable excited state of HeH^+ and not to the ground state. While not directly related to the subject of antimatter production, this investigation did shed a great deal of light on the recombination process in general and so is worthy of inclusion in this report. A paper describing this work has been accepted for publication and is included as appendix A.

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B. REFERENCES

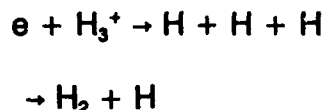
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CHAPTER 2

ELECTRIC FIELD EFFECTS ON THE DISSOCIATIVE RECOMBINATION OF H_3^+

A. INTRODUCTION

The dissociative recombination of H_3^+ , i.e.:



has proved to be an enigmatic problem that has received much attention over the last ten years. It is an important problem since H_3^+ is usually the dominant ion in molecular hydrogen plasmas that are found in interstellar clouds, planetary ionospheres, fusion edge plasmas and negative ion sources^{1,2}. H_3^+ will also be an important intermediate ion in any scheme for the stepwise production of antihydrogen cluster ions.

Prior to 1984, several experimental groups³⁻⁸ that had studied H_3^+ recombination came to the consensus that the room temperature rate coefficient had a value of about $2 \times 10^{-7} \text{ cm}^3\text{s}^{-1}$. In 1983 however, Adams et al.⁹ published the results of a FALP (Flowing Afterglow Langmuir Probe) experiment that indicated that the rate was in fact much smaller, ($< 2 \times 10^{-8} \text{ cm}^3\text{s}^{-1}$). They argued that their experiment applied to ground vibrational state ions while the higher rates measured in other experiments could be explained in terms of vibrationally excited ion recombination, influencing the data. A merged beams experiment¹⁰ addressed this point and indeed did show that when vibrationally cold ions were used, a low cross section corresponding to a 300K rate coefficient of about $2 \times 10^{-8} \text{ cm}^3\text{s}^{-1}$ was found. One problem with this finding was that it

contradicted an earlier inclined beam experiment⁵ that used vibrationally cool ions and that found large recombination cross sections.

Throughout the decade of the 80's, the controversy raged as more experiments were performed. Amano^{11,12} used an infra-red laser absorption technique and found a rate coefficient of $1.8 \times 10^{-7} \text{ cm}^3\text{s}^{-1}$ for ions that were well defined to be in the $v=0$ level. Canosa et al.¹³ used a modified FALP apparatus that used a moveable mass spectrometer to monitor the decay of the H_3^+ ions directly and found a 650K value of $1.1 \times 10^{-7} \text{ cm}^3\text{s}^{-1}$ for vibrationally cold ions. In addition, an analysis of the quenching of vibrationally excited states of H_3^+ ions in FALP afterglows by these authors indicated that the ions used in the experiment of Adams et al. were not in fact in their ground state as previously surmised. Following an extensive re-measurement of the rate using a similar FALP apparatus, Smith and Spaniel¹⁴ still claim that the low value of $2 \times 10^{-8} \text{ cm}^3\text{s}^{-1}$ applies.

The original merged beams experiment of Hus et al.¹⁰ has been repeated in our laboratory a number of times using a different apparatus and a different ion source and the result of a low measured cross section for cold ions has each time been confirmed. A different merged beams experiment, using the electron cooler at the CRYRING storage ring has been performed in Stockholm¹⁵ and following re-analysis of their initially published results¹⁶ they find a cross section of $1.43 \times 10^{-16}/E^{1.15} \text{ cm}^2$ (at low center-of-mass energy) that corresponds to a room temperature rate of $1.15 \times 10^{-7} \text{ cm}^3\text{s}^{-1}$.

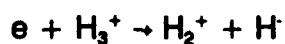
Underlying all of this controversy is the problem that the absence of an appropriate curve crossing between the H_3^+ and H_3^* systems means that vibrationally cold H_3^+ ions

cannot undergo direct dissociative recombination^{17,18}. A simple theoretical argument therefore would predict a very small reaction rate. Mitchell and Rowe¹⁹ addressed this problem by suggesting alternative recombination paths such as a capture involving a large change in vibrational quantum number to a broader potential energy curve that could be predissociated or via a capture into a high Rydberg state of the molecular ion that was stabilized by an unspecified predissociation. Unusually high predissociation rates for high Rydberg states have been observed experimentally in other studies²⁰⁻²² of H_3^+ .

Bates²³⁻²⁵ proposed a multi-step mechanism involving an initial capture into a vibrationally excited, autoionizing Rydberg state of H_3^+ followed by a series of radiationless transitions to lower n , higher v states of the molecule. He argued that such a mechanism could in fact provide for the rapid recombination of the ion with thermal electrons.

The current authors have devised a number of experiments to test the hypothesis that the low cross sections found in their merged beam experiments are due to the destruction of high Rydberg intermediate states by the electric fields used to control the beams in their apparatus. An earlier study of the branching ratio of the product channels revealed that a third channel consisting of long-lived ($> 10^{-7}$ s) H_3^+ molecules was detected following the recombination of ground state H_3^+ ions²⁶. Such a channel would of course be an intermediate state in the multi-step recombination mechanism.

In a recent measurement, Yousif et al.²⁷ have examined the ion pair formation reaction:



that is a minority recombination channel. This reaction is endothermic for ground state ions, displaying an energy threshold at 5 eV. It also proceeds via a direct transition from the ion to a diabatic state of H_3^{**} that intersects the ion state in the vicinity of the $v=4$ level^{17,18}. The important thing to note about this channel is that unlike the recombination to neutral channels it does not involve the formation of high Rydberg states of the neutral molecule. Yousif et al. found that their results agreed closely with an earlier inclined beam measurement of Peart et al.²⁸ despite the fact that similar measurements of the recombination to the neutral channels by the two groups^{5,10} differed by over an order of magnitude at low energies¹ and by two orders at energies above 0.5 eV!

The present paper describes an experiment that has been performed in which the electric field experienced by the newly recombined H_3 system is systematically lowered and effects of this on the measured signal count rate have been observed.

B. EXPERIMENTAL

Figure 2.1 shows a schematic of the experimental arrangement used for this study. The MEIBE II apparatus at the University of Western Ontario was used and this device incorporates a pair of field ionization detectors that have been used to detect excited atoms formed following electron-ion recombination^{29,30}. These devices are not used *per se* in this experiment except to act as very effective collimators that prevent scattering of the primary ion beam into the neutral detector as the electric field of the analyzer A is reduced from its normal operating value of 3.0 kV/cm to a value of 220 V/cm.

¹ Based upon an extrapolation of the inclined beam results to 10 meV.

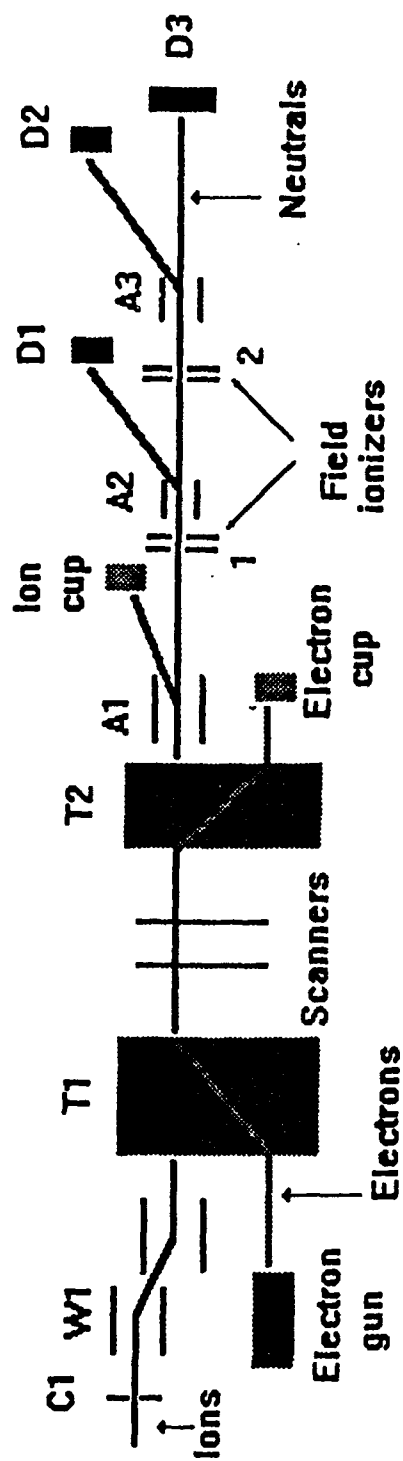


Figure 2.1
Schematic diagram of the MEIBE II apparatus, configured for field ionization studies.

The ion beam is formed in a high pressure radio-frequency ion source mounted in the terminal of a 400 KeV Van de Graaff accelerator. This source has been demonstrated by us to be capable of producing ground vibrational state ions. The ions are accelerated, mass analyzed and passed into the interaction region illustrated in the figure. The electron beam is formed from an indirectly heated barium oxide cathode and accelerated so that the electrons have a velocity close to that of the ion beam. The beam is confined spatially using a 25 Gauss axial magnetic field. A transverse electric field of 200 V/cm is applied at T1 and this causes the electron beam to perform a helical motion inside this so-called trochoidal analyzer. As the beam leaves this device, it has made one complete helical rotation and now travels along the field lines again, but now co-axial with the ion beam. The two beams interact over a distance of 8.3 cm before the electrons are separated from the ions using a second trochoidal analyzer T2 again with an electric field of 200 V/cm. The ion beam passes through T2 and is then deflected by A1 into a Faraday cup where its current is measured. The electron beam is collected in a second Faraday cup.

The neutrals formed in the interaction region pass undeflected and are counted using a surface barrier detector. Those formed due to beam-background gas collisions are distinguished from those due to electron-ion collisions by modulating the electron beam and counting in and out of phase with the chopping frequency. The true signal is determined from the difference in these two counts.

As mentioned above, the aim of this experiment was to monitor the signal count rate from the recombination of electrons with vibrationally cold H_3^+ ions as the electric

field, experienced by the neutrals was systematically reduced. In practice, the field on analyzer A1 was reduced while that on the trochoidal analyzer was left unchanged. As A1 was reduced, the ion beam moved out of the Faraday cup and so was not measured directly. The current picked up by collimator C1 at the entrance to the interaction region was monitored however, during the measurements so that the true ion current could be calibrated. The small apertures in the field ionizers do intersect some of the neutrals and so the cross sections measured in the experiment can not be considered absolute. Since we are looking for a relative effects however, this is not critical and we can normalize the measurements to previous, absolute measurements taken without the restrictive apertures.

C. RESULTS

Figure 2.2 shows the measured recombination signal as a function of A1 analyzer electric field. It is seen that there is a striking increase in this signal as the field is decreased indicating that high Rydberg states must be being formed in the recombination process and subsequently field-ionized in the analyzer field typically used in this apparatus. For ground vibrational state H_3^+ and low energy electrons, the H and H_2 recombination products can only be formed in their ground electronic states so this finding means that the species that are being ionized are H_3^+ molecules! Figure 2.3 shows the signal with the abscissa labelled in terms of the n value that will be ionized by the field setting at which the data point was taken. This n value has been calculated using the equation³¹:

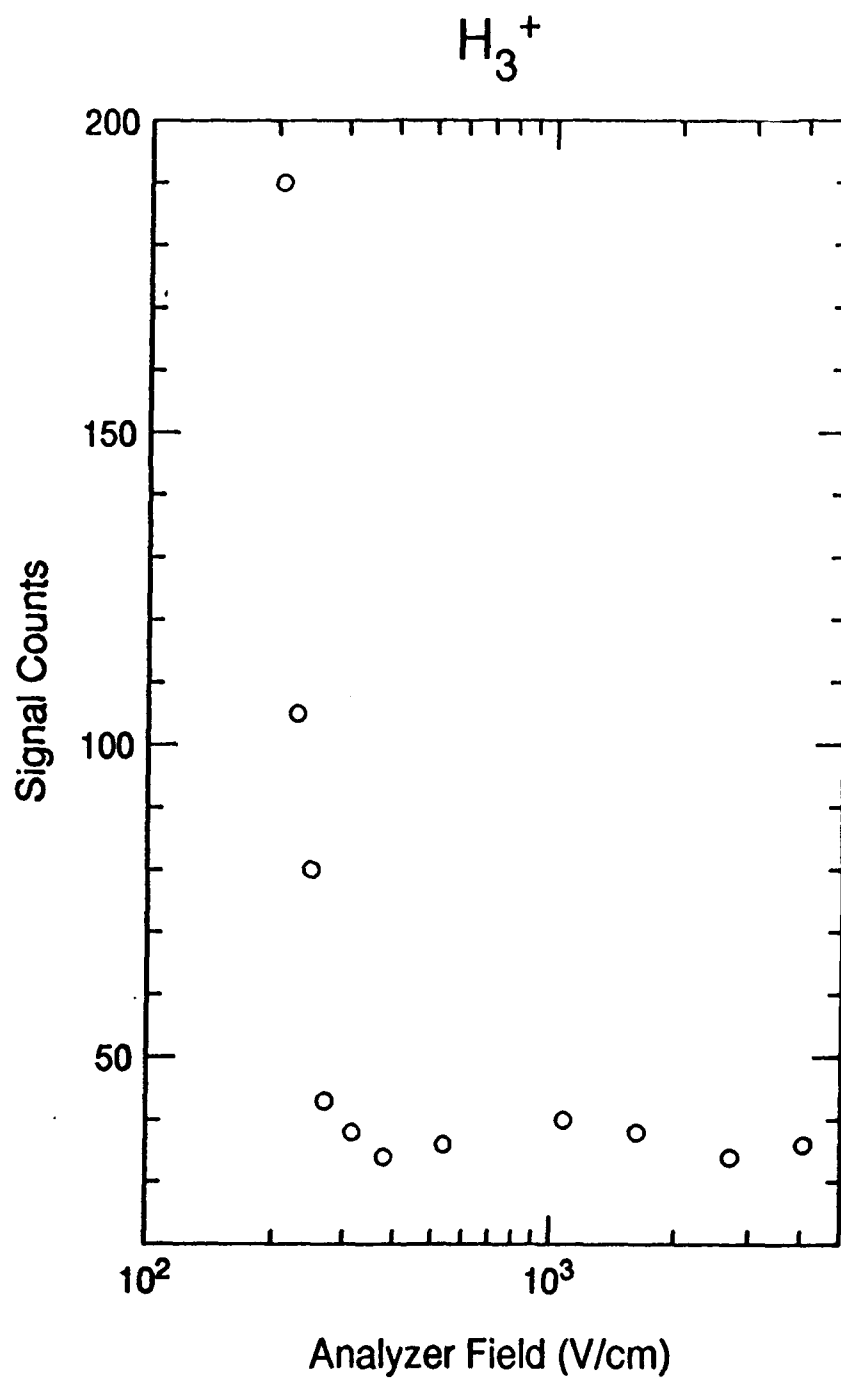


Figure 2.2
Plot of signal counts versus analyzer field.

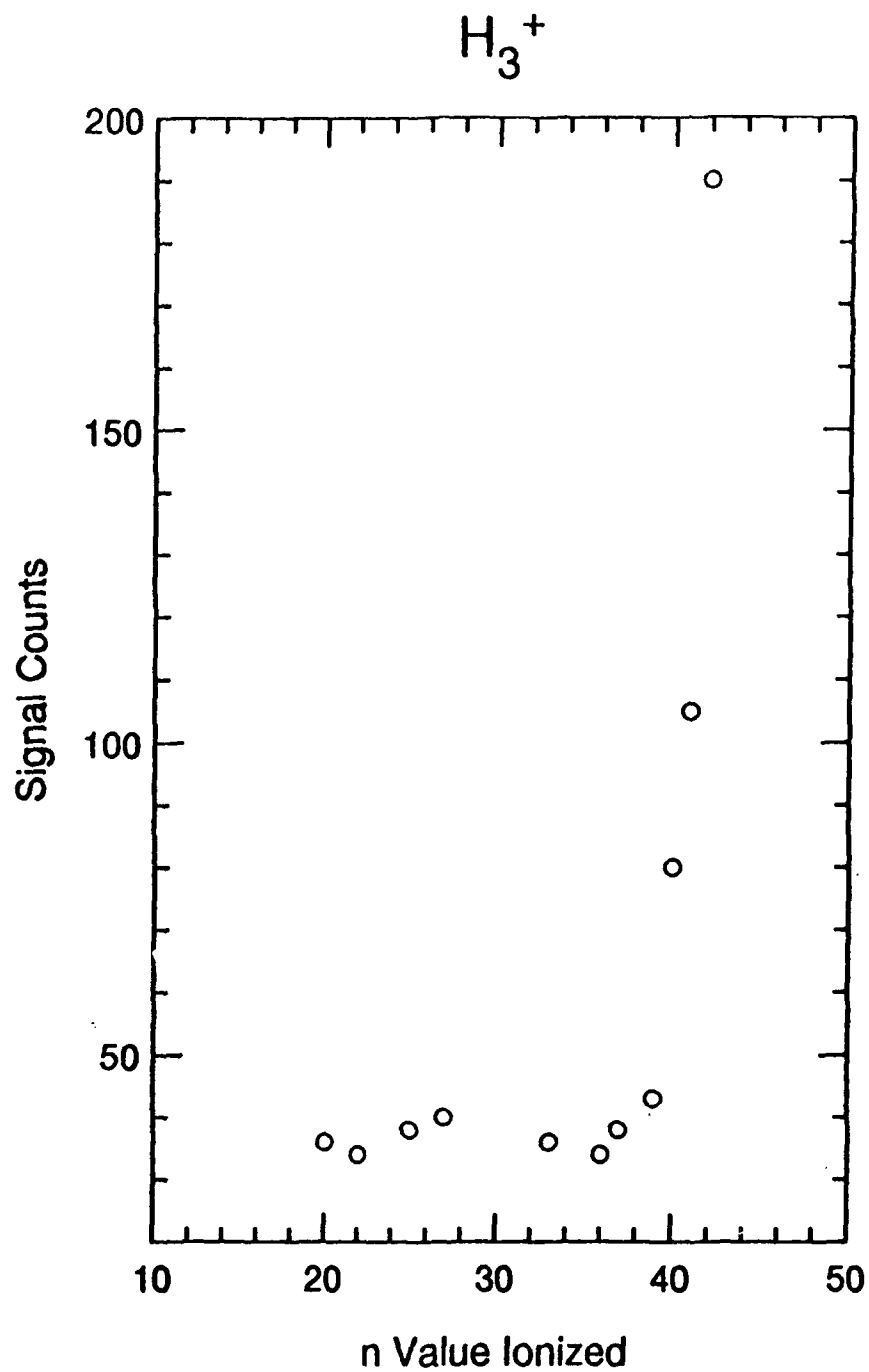


Figure 2.3
Plot of signal counts as a function of n value field ionized in the analyzer.

$$E_{crit} = \frac{6.4 \times 10^8}{n^4} \text{ Vcm}^{-1}$$

It is seen that the large increase in the signal observed when the field is lowered below 400 V/cm can be attributable to the survival of molecules with principal quantum numbers greater than 36.

Figure 2.4 shows the signals (measured with fields of 3.0 kV/cm and 206 V/cm) as a function of Center-of-Mass Energy and it is seen that they are well behaved, falling off as expected with increasing energy with different slopes reflecting the multiplicative factor between them². This is important for it was found that at lower electric field strengths (155 V/cm), where the ion beam is barely deflected, spuriously large signals caused by space charge interaction between the electron and ion beams were produced. These signals are also shown on the diagram and it can be seen that they have an anomalous energy behaviour. Distortion of the pulse height spectrum from the surface barrier detector was also seen with the full energy signal peak (due to H+H+H, H₂+H or H₃⁺) becoming dominant⁶. Care was taken to ensure that the signals presented here were real and that the cross sections determined from these signals were independent of electron beam currents. Space charge induced signals should vary with the beam current). This is illustrated in Table I and it is seen that only at the lowest field do such problems arise.

Figure 2.5 shows the measured cross sections for H₃⁺ recombination measured with low analyzer field strength (220 V/cm) and with the normal field strength

² Space charge induced signals would be constant over the energy range and would yield an additive relationship between the two sets of signals.

H_3^+ Recombination

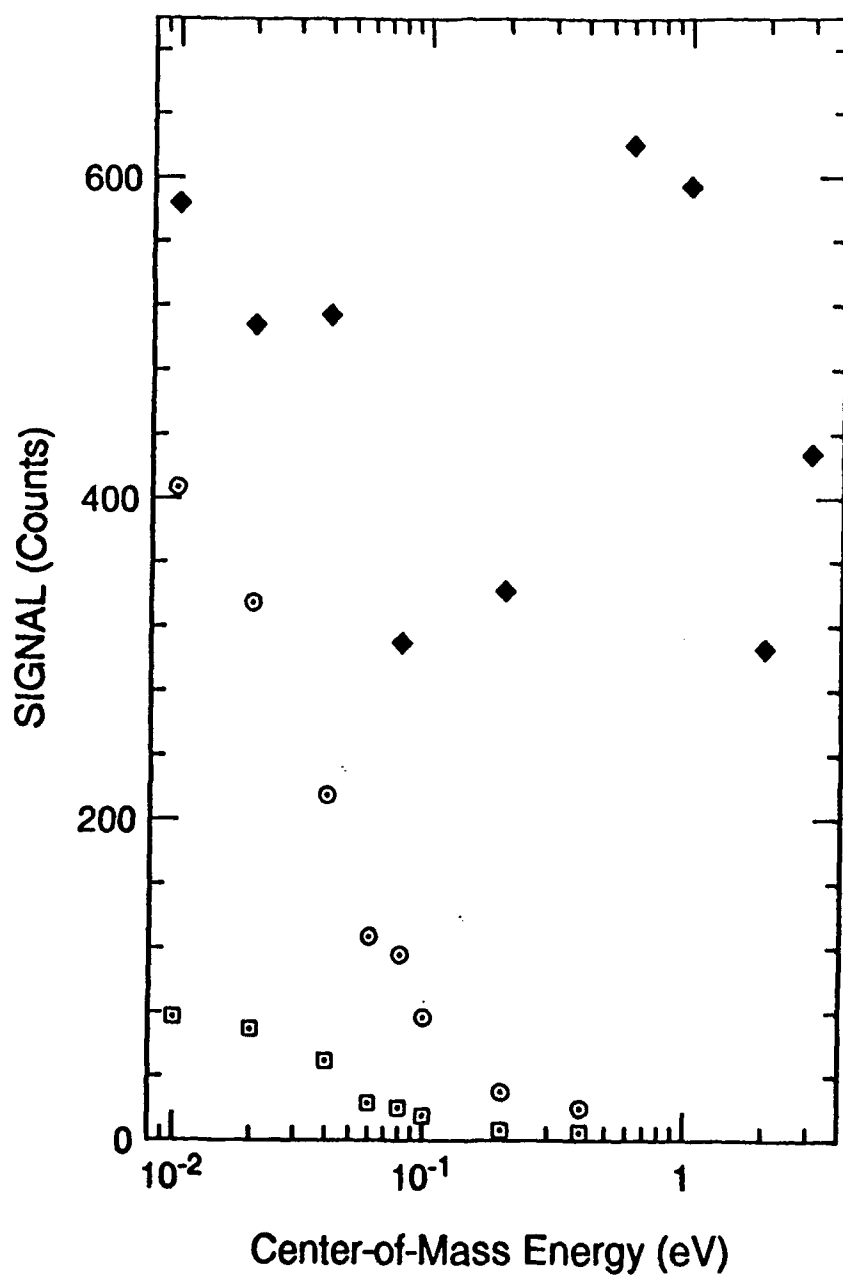


Figure 2.4

Plot of signal counts versus center-of-mass collision energy for different analyzer field settings. Open squares; 3kV/cm. Open circles; 206 V/cm. Diamonds; 184 V/cm.

H_3^+ Recombination

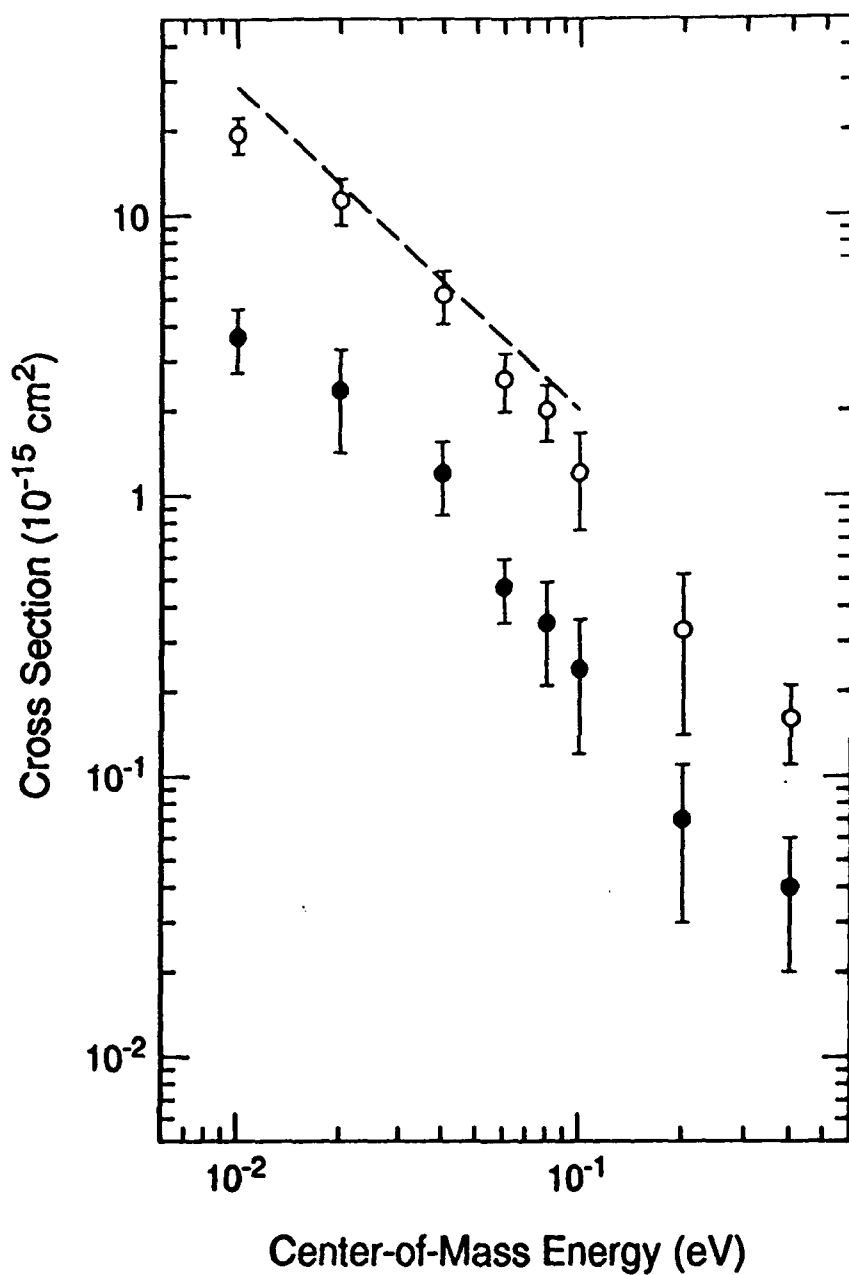


Figure 2.5

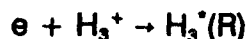
Cross sections for the dissociative recombination of H_3^+ measured with an analyzer field of 220 V/cm (open circles) and 3 kV/cm (closed circles). Dashed line represents results taken at CRYRING in Stockholm¹⁶.

(3.0 kV/cm). A factor of five increase is seen over the measured energy range when the field strength is reduced. The dashed line shown in the figure represents the cross sections recently published by the Stockholm storage ring group¹⁶ and it is seen that there is excellent agreement between our measurements and theirs. As in the present results, the storage ring measurements also exhibited a more rapid fall-off above 0.1 eV¹⁰ but this is not shown. The Stockholm results correspond to a rate coefficient of 1.15×10^{-7} cm³s⁻¹ at 300K in good agreement with afterglow measurements of Canosa et al.¹³, Amano et al.^{11,12} and MacDonald et al.⁴ and so our revised results agree with these authors also.

Analyzer Field (V/cm)	Electron Current (μ A)	Cross Section (10^{-15} cm ²)
238	46	2.9
	30	3.25
	18	3.05
210	46	2.8
	30	2.75
	18	2.45
188	46	3.2
	30	3.6
	18	4.35
184	46	0.25
	30	1.2
	18	4

D. DISCUSSION

If we consider an electron with 10 meV of kinetic energy colliding with the $v=0$ level of the H_3^+ ground state, the indirect capture process:



must proceed to a ro-vibrationally excited state of the underlying Rydberg state. For this to occur the transition must proceed via a $\Delta v = +1$ transition to a Rydberg state with an n value of about 7 or via a transition to a rotationally excited state with a high n value. Takagi³² has discussed the role of rotational excitation in the recombination and found it to be significant and so one can expect that the latter process must be important. Bates multi-step mechanism²³⁻²⁵ can subsequently be invoked to explain how these states proceeds to predissociation and subsequent stabilization.

One of the key questions here concerns the lifetime of the intermediate Rydberg state. In order for the capture to be rapid, the autoionization rate for the initial state must be high. Why then can it survive for long enough for the multi-step process to effect stabilization? The answer to this may lie in the availability of many ro-vibrational modes of the polyatomic system that can inhibit the autoionization process.

In our apparatus, the recombination takes place in a region where the electric field is extremely small. It is only after the recombined products have travelled through the second trochoidal analyzer that they experience the high fields in the ion beam analyzer. Given that the energy of the beam is 350 KeV and that the distance between the end of the 8.3 cm long interaction region and the analyzer entrance is 6 centimeters, the lifetime of the state that becomes field ionized must be in the range 12-30 ns or greater.

Once in the analyzer field, the particular state into which the electron has been captured will undergo Stark broadening and can mix with other Stark broadened states. Bordas and Helm²¹ have recently presented an analysis of the field ionization of Rydberg state of H_3 and we shall follow their treatment in the following discussion. If formed, an $n=7, v=1$ state could mix with high $n, v=0$ states. For the case of a 3.0 kV/cm field, the Stark splitting of the $n=7$ level is very small, ($\Delta E_B \sim 0.0025$ eV) while that of, for example, the $n=35$ state is substantial ($\Delta E_B \sim 0.08$ eV). Figure 2.6 shows the ionization thresholds for the uppermost and lowest Stark components (the so-called "red" and "blue" states) and it can be seen that at this field strength all Stark components of $n=35$ will be field ionized. Thus in this case, the presence of the electric field will give rise to a reduction in the measured recombination signal.

If the field experienced by the neutrals was extremely high, it may be possible to obtain enhanced stabilization in addition to field ionization due to the Stark splitting, as the lower Stark components of the $n=7$ state are pushed down in energy below the ionization potential. This may explain why similar electric field effects have not been seen in the Stockholm measurements where the field experienced in the first magnetic deflector following the interaction region is of the order of 500 kV/cm. This field strength is sufficient to directly field ionize the upper red sub-states but not the lower blue sub-states. At this field strength, the lower blue components of the $n=7, v=1$ state can be suppressed by as much as 0.15 eV, bringing them well below the ionization threshold of the $H_3^+ v=0$ state and allowing mixing with Stark components of stable lower n states. Thus we have the unusual situation of the field enhancement of recombination.

This argument is of course speculative and is presented in an attempt to explain differences in results from different experiments. It does perhaps provide a starting off point in our attempt to understand this perplexing problem of H_3^+ recombination. If the argument is valid however, it can also perhaps be used to explain the differences found in afterglow measurements since under the conditions encountered in these experiments, the high lying long lived states can undergo collisional mixing with competition arising between stabilization and reionization.

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CHAPTER 3

LASER STIMULATED RADIATIVE RECOMBINATION OF He^+

A. INTRODUCTION

In our previous report¹ we reported the first measurements of the stimulated radiative recombination of He^+ ions with electrons in the presence of a radiation field from a CO_2 laser. Two double peaked structures were found and these were attributed to capture into the $n=15$ and $n=16$ states. In fact due to an error in our analysis, these structure should have been assigned to the $n=11$ and $n=12$ levels of He. At the time it was also proposed that the double peaking was due to capture into the singlet and triplet manifolds of helium. A subsequent examination of the energy spacing of these manifolds indicated that in fact the structures could also be due to capture into different l states. While these are degenerate for hydrogen, and therefore only single peaks appear in the stimulated recombination spectrum, different l states are non-degenerate in helium and so multi-peaked structures would be expected. A search has been initiated for other associated peaks and the initial results for the $n=11$ level are shown in figure 3.1. It should be emphasized that this study is not complete but it is seen that in fact five peaks have been found in the cross section spectrum.

Figure 3.2 shows the total collected data so far with the $n=11$ and $n=12$ peaks identified.

He Stimulated Recombination

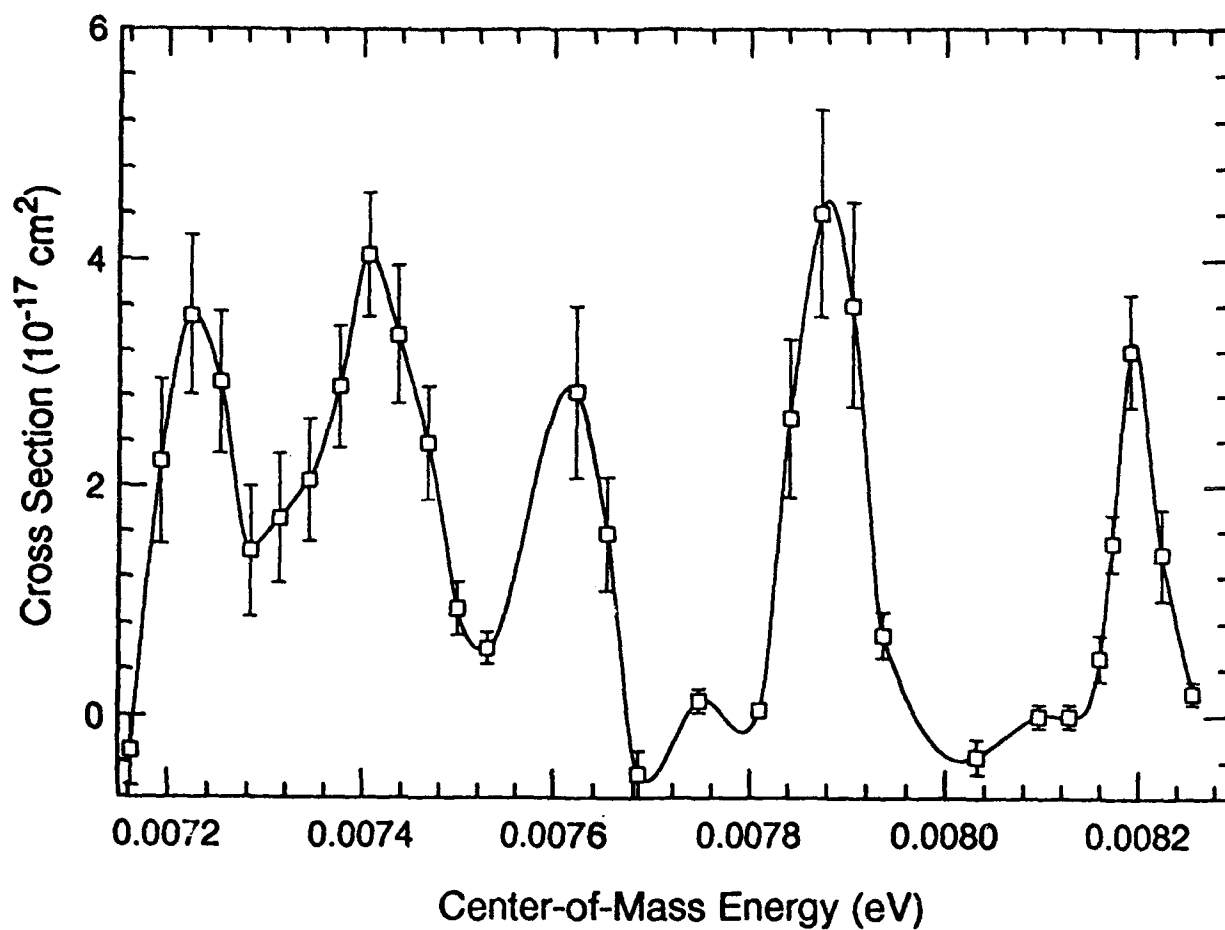


Figure 3.1

Effective cross sections for stimulated radiative recombination of He⁺. The structures are associated with capture to the n=11 states of helium.

He Stimulated Recombination

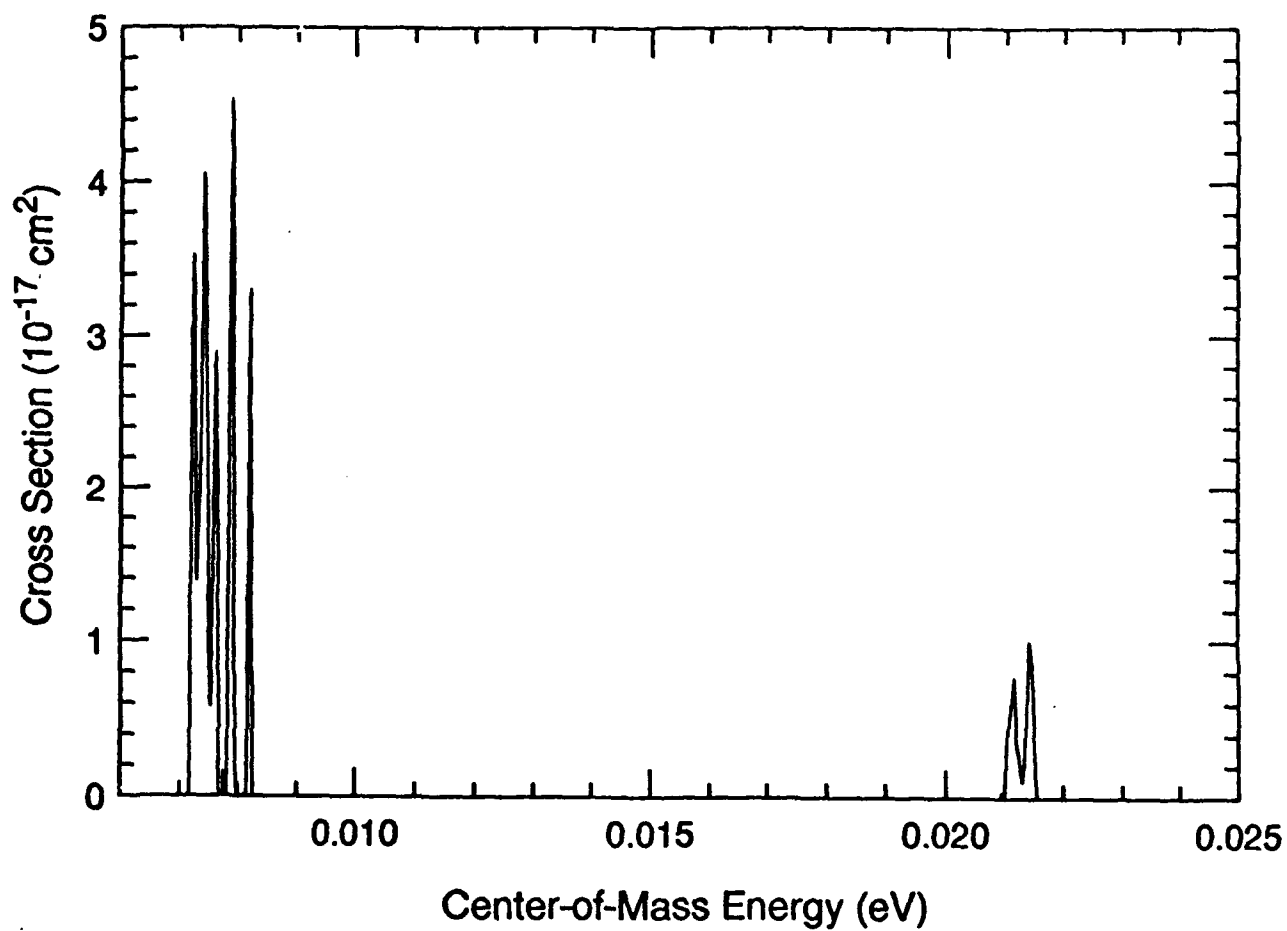


Figure 3.2

Effective cross sections for stimulated radiative recombination of He⁺. The structures are associated with capture to the n=11 and n=12 states of helium.

B. REFERENCES

1. *Merged Beam Studies of Laser Stimulated Radiative Recombination: 4th Report. 30 June 1993, J.B.A. Mitchell*

APPENDIX A

DISSOCIATIVE RECOMBINATION OF HeH^+ : A RE-EXAMINATION

(To appear in Physical Review A. June 1994)

DISSOCIATIVE RECOMBINATION OF HeH^+ : A RE-EXAMINATION

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ABSTRACT

A high energy resolution study of the dissociative recombination of HeH^+ has been performed. Theoretical analysis has indicated that the recombination at low energy is due to the presence of a metastable triplet state component of the ion beam.

Physics Abstracts Classification No: 34.80 Kw

INTRODUCTION

In a previous study¹, a large cross section was found for the dissociative recombination (DR) of HeH^+ at thermal energies. Numerous calculations²⁻⁶ of the potential-energy curves for HeH and HeH^+ have shown, however, that the ground ($X\ ^1\Sigma^+$) electronic state of HeH^+ , does not have a suitable curve crossing with a repulsive neutral state through which the dissociative stabilization of the recombination could proceed. A second measurement of the recombination of HeH^+ using an ion-storage ring has recently been reported⁷ and again a large cross section was found at low center-of-mass collision energies. This measurement also revealed the presence of a peak in the cross section in the vicinity of 20 eV. Recent theoretical calculations⁸ of the elastic scattering of electrons from ground state HeH^+ indicated the presence of resonances that coincide with the positions of resonances found in reference 1. Given these developments, a decision was made to re-examine this process using our merged beams apparatus at the University of Western Ontario. These measurements are described in this paper. We have, however, produced a re-analysis of this data that has indicated that the observed large cross-sections arise not from the recombination of the ion ground state but in fact from a metastable triplet state of the ion. This explanation of the observed recombination was originally suggested by Michels².

EXPERIMENTAL TECHNIQUE

The present experiment has been performed using the MEIBE II merged beams apparatus at the University of Western Ontario. The ions are formed in a high pressure radiofrequency ion source moun-

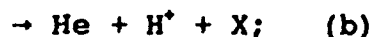
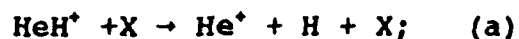
ted in the terminal of a 400 keV Van de Graaff accelerator. The previous experiment employed a radiofrequency trap ion source that delivered very small beam currents and having an electron filament, was technically more difficult to operate in the Van de Graaff terminal. We have modified a conventional radiofrequency ion source by decreasing the diameter of the extraction canal from 1 mm to 0.5 mm. This has allowed the source to be operated at higher pressures*. During the first series of measurements reported here, the source operated with a source gas consisting of 50% hydrogen, 50% helium at a pressure of between 20 and 30 mTorr. As seen below the recombination cross section and the positions of resonances obtained using ions derived from this source agree very closely with those obtained from the previous work.

Following acceleration, the ions are mass analyzed and injected into the merged beams apparatus. Upon entering this apparatus, they are deflected twice to remove any neutrals formed in transit and then enter the interaction chamber. In this region, the electron beam is formed from an indirectly-heated barium oxide cathode and merged with the ion beam using a trochoidal analyzer. After interacting over a distance of 8.6 cm, the electron beam is de-merged from the ions and collected in a Faraday cup. The ion beam is electrostatically deflected into a second Faraday cup while the neutrals pass on undeflected to a surface barrier detector. Others details of the apparatus have been published elsewhere¹⁰ and the method for determination of the cross section is the same as in

* This idea was first used by Knystautas and Lapointe⁹ for the production of beams of multiply-charged atomic ions from r.f. sources.

ref.1.

The HeH^+ ions undergo the processes:



in collision with a background gas molecules, X, and electron-ion dissociative recombination:



in collision with the electron beam. Neutrals arising from background gas collisions (a,b,c) are separated from those formed by electron-ion interactions (d) by modulating the electron beam and counting the particles in and out of phase with the modulation. The difference in the two counts yields the true electron-ion signal.

RESULTS

Measured cross sections for the dissociative recombination of HeH^+ , over the energy range from 0.01 eV to 22 eV are shown in figure 1. The low energy region is shown with an expanded scale in figure 2. Comparison of these figures and the previous measurement¹ show that they are almost identical at low energies. The present results have more clearly defined resonant structures than demonstrated in the previous work where the energy spacing used was larger and where the region above 0.1 eV was not investigated in detail. A peak is found in the vicinity of 20 eV as in the case of the storage-ring measurements⁷. In fact closer examination shows this to consist of a multiple peaked structure. (See Figures 5 a and b).

Given the fact that there is no suitable curve-crossing for ground state HeH^+ ions, we decided to see if the low energy results could possibly be explained in terms of the recombination of excited state ions*. The lowest electronically excited states of HeH^+ arise from the atomic helium ion $\text{He}^+(1s)$ and the neutral hydrogen atom, $\text{H}(1s)$. At $R=\infty$ the excitation energy is equal to the difference between the helium and hydrogen atomic ionization potentials: $I_{\text{He}} - I_{\text{H}} = 10.98$ eV. The total two-electron spins of these states may be equal to 1 and 0 and so we have two energetically different states $a^3\Sigma^+$ and $A^1\Sigma^+$ which are degenerate at $R=\infty$. The energy curves of these states were calculated in ref.3. The potential energy curves of both states are repulsive at small internuclear distances R but weak attractive potential wells are created at large R by polarization and exchange forces, (Fig. 3). The $a^3\Sigma^+$ state has a little more the binding energy at all R than $A^1\Sigma^+$.

Since the $a^3\Sigma^+$ state of HeH^+ has a total two-electron spin equal to unity, ($S=1$), direct radiative transition to the ground $X^1\Sigma^+$ state, which has $S=0$, is forbidden by the spin conservation rule. This transition may occur only by weak spin-orbit interaction but to a first approximation, the rate for this transition is zero for Σ states of diatomic molecules. The relativistic magnetic dipole transition is the predominant means of decay¹¹ of the 2^3S state of

* In Ref. 1, the dissociative excitation measurements seemed to indicate that the HeH^+ ions were in their ground electronic and vibrational states. We now believe that these results were a manifestation of a strong resonant process associated with the ground state but that they did not preclude the existence of excited states in the beam.

He. If this is also true for the $a^3\Sigma^+$ of HeH^+ , the rate for this decay might be expected to be about a factor of 100 slower than for $\text{He}(2^3S, \tau \sim 8000 \text{ s})$ since the overlap between the initial and final electronic wavefunctions is so poor. The transition $A^1\Sigma^+ \rightarrow X^1\Sigma^+$ is optically allowed but may also be slow because of the poor overlap of the electronic wavefunctions of these two states. The transit time between the ion source and the interaction region is $\sim 1.2 \mu\text{s}$ and if the lifetime of the $A^1\Sigma^+$ state exceeds this, it may also be present in the ion beam. The existence of metastable excited ions in HeH^+ beams has already been established by Schopman and Los¹²

The degeneracy at $R=\infty$ of the triplet $a^3\Sigma^+$ and singlet $A^1\Sigma^+$ energies levels plays an important role for the recombination of the metastable molecular ion. Let us consider the neutral HeH^{**} state (figure 4) which consists of the singlet $^1\Sigma^+$ molecular core and a Rydberg electron in a state (n,l) . In a zero order approximation, the energy of the neutral HeH^{**} complex, E_0 may be written as the sum $E_0 = E_{\text{core}}^1 + \epsilon(n,l)$ where E_{core}^1 is the energy of the core molecular ion in the singlet $^1\Sigma$ state and the energy of the Rydberg electron $\epsilon(n,l)$, is equal to $-1/2n^2$,

i.e.
$$E_0 = E_{\text{core}}^1 - 1/2n^2$$

These states can decay to produce the triplet $a^3\Sigma^+$ state with the outgoing electron having a small kinetic energy if the internuclear separations are to the left of the stabilization points (figure 4). These states can also autoionize leading to the release of a fast electron ($\sim 10 \text{ eV}$), leaving the ground molecular ion complex $\text{He}(1s^2) + \text{H}^+$ which dissociates. We can see from figure 4 that the energy levels with $n=7,8,\dots,16$ cross the range of the ground

vibrational level of the triplet molecular ion, i.e. the $a^3\Sigma^+$ state has many crossings suitable for DR. It should be noted that the total spin of the system $e+\{\text{HeH}^+, ^3\Sigma\}$ may be equal to $3/2$ and $1/2$ and so the quartet states may also have crossings with the $a^3\Sigma^+$ state. The singlet state may also be present in the beam but it will not contribute to the recombination. It is the term limit of the repulsive states shown in figure 4 and so there is no intersection with these states. Transition to the quartet manifold is not possible due to spin conservation.

In order to see if there is any evidence for the $a^3\Sigma^+$ state contributing to the recombination, it was decided to calculate the vibrational levels of this state. To do this one must solve the Shrodinger wave equation:

$$\frac{d^2\phi}{dR^2} + \frac{2\mu}{h^2} [E - V(R)] \phi = 0; \dots (1)$$

where V are the potentials shown in fig.3; μ is the reduced mass of the molecular ion: $\mu = M_1 * M_2 / (M_1 + M_2)$; ϕ is the wave function of the vibrational motion. In the semiclassical approximation these levels may be determined by means of the Bohr-Sommerfeld quantization rule:

$$\int_{R_1}^{R_2} [2\mu (E - V(R))] dR = \pi h (v + 1/2); \dots (2)$$

where v , ($=0, 1, 2, \dots$) is the vibrational quantum number and R_1, R_2 are the turning points. In order to estimate the total number of vibrational levels supported in a given well, let us consider $E=0$ in (2) and compute the integral in the range $R_0 < R < \infty$ where

$R (=4.5 a_0)$ is the equilibrium point of the potential well. In this range, the potential $V(R)$ may be represented as the polarization potential $V(R) \approx -\alpha/2R^4$ where $\alpha=4.5$ is the polarizability (in atomic units) of the hydrogen atom in the ground state. From this calculation, we have obtained

$$N_{\max} = v_{\max} + 1$$

for the maximum number N_{\max} of vibrational levels, where:

$$v_{\max} = \frac{\sqrt{\mu \alpha}}{\pi R_0} - \frac{1}{2};$$

The values of N_{\max} for $a^3\Sigma^+$ are given in table 1a for all the isotopomers of HeH^+ and in table 1b for the $A^1\Sigma^+$ state of $^4\text{HeH}^+$.

The energies of the vibrational levels were determined quantum mechanically by solving equation (1) as the usual eigenvalue problem. The problem was also approached semi-classically. The integral in (2) was calculated numerically as a function of energy E and the levels were determined as roots of equation (2). The results for the $a^3\Sigma^+$ and $A^1\Sigma^+$ states are given in Tables 1 a,b. It was found that the semiclassical and quantum mechanical approximations give the same results. The reason for this is that the potential V and its gradient in (1) are small and therefore the semiclassical approximation is excellent in the case of the $a^3\Sigma^+$ molecular state.

If we compare the energy differences ΔE_{0v} between the calculated 0 and v vibrational levels:

$$\Delta E_{01} = 0.0372 \text{ eV}; \quad \Delta E_{02} = 0.0612 \text{ eV}; \quad \Delta E_{03} = 0.0748 \text{ eV}; \quad (4)$$

(indicated by the arrows in figure 2), we see that these values are in excellent agreement with the positions of the experimental

resonances. This agreement suggests that the recombination observed in this experiment is actually associated with the metastable $a^3\Sigma^+$ state, the resonances arising from competition between the recombination and vibrational excitation¹³⁻¹⁶. Furthermore, the large depth of the resonances indicates that the ions were predominantly in the $v=0$ level of this state.

The disappearance of the cross section in the energy range between 0.55 and 0.90 eV may be explained by examination of figure 4. It can be seen that transitions from the initial ion state to the repulsive dissociating states can occur for center-of-mass energies less than 0.55 eV but not for higher energies. The 20 eV energy peak in the cross section may be explained in terms of electron capture from the ground state of the HeH^+ ion to a repulsive doubly-excited state such as those illustrated in figure 4. The reasons for the dips between 0.09 and 0.55 eV and for the peak between 0.9 and 1.2 eV are not clear and are perhaps associated with some other excited ion state or recombination process. Initial calculations show that the positions of these resonances correlate with transitions involving the second triplet state of HeH^+ . In the earlier studies of Yousif and Mitchell¹, the dissociative excitation results gave an indication that this state was populated under low pressure source conditions. It is not clear however, how this state can survive passage from the source to the collision region for it can make radiative transitions to the first triplet state. A more careful examination of the effects of rotational excitation on the lifetime of this state is currently underway to see whether this might be a reason for its survival

into the interaction region. The results of this study will be published later and a discussion of the associated resonances will be deferred until then.

Having obtained convincing theoretical evidence for the hypothesis that the recombination observed at low energies was actually associated with the $a^3\Sigma^+$ state of the ion, a method was sought for experimental verification of this hypothesis. One way to remove spin-forbidden metastable states from an ion beam is to introduce oxygen into the ion source. Oxygen has a lone pair of electrons and electron exchange reactions can occur between the O_2 molecule and the ion resulting in the quenching of the excited states¹⁷. This technique had been demonstrated by us in previous studies of CH^+ recombination¹⁸. In that case it was found that addition of oxygen to the source gas resulted in the decrease of the CH^+ recombination cross section because the original ion beam contained a fraction of the ions in the $^3\Pi$ excited state.

The results shown in figure 1 were taken using a 50:50 H_2 :He gas mixture in the ion source. When this was replaced with a 25:25:50 H_2 :He: O_2 mixture, the low energy cross sections were found to decrease to undetectable levels as shown by the solid circles in figure 2. This is regarded as convincing proof that the observed signal was in fact due to the recombination of the $a^3\Sigma^+$ state of HeH^+ and not to the ground singlet state. Further evidence for the presence of a large excited state component in the ion beam is provided by the change in the 20 eV peak with the addition of oxygen to the source. This is illustrated in figures 5a and b where it is seen that these peaks are enhanced by about a factor of two

by the use of oxygen. The 20 eV peak structure is due to recombination arising from transitions from the ground state of HeH^+ to upper, dissociating, neutral Rydberg states such as those shown in figure 4 and if a substantial fraction of the beam is excited, then a large enhancement of this cross section would be expected when the excited states were quenched. The fact that there is an enhancement by about a factor of two suggests that perhaps 50% of the beam used for the original measurements consisted of ions in excited states. This would mean that the low energy cross sections shown in figures 1 and 2 should actually be multiplied by a factor of two, since they arise from this excited state component.

DISCUSSION

When the original measurements of HeH^+ recombination were first made by Yousif and Mitchell¹, it was a surprise to find that the measured cross section was so large given that there did not seem to be any obvious mechanism through which the recombination should proceed rapidly. The present work provides an explanation for this, based upon the recombination of excited state ions that is supported by both theoretical modelling and experimental observation. Having said that it should be noted that there has been a recent theoretical study by Sarpal et al.¹⁹ of the dissociative recombination of ground state HeH^+ ions that agrees well the earlier results. This calculation invoked indirect recombination through the neutral Rydberg manifold lying below the HeH^+ ground state.

HeH^+ is not of course the only species whose recombination has

been the subject of controversy. H_3^+ also displays a rapid recombination rate despite the presence of a suitably placed curve crossing. Bates et al.²⁰ has recently discussed the theoretical situation and has proposed a multistep mechanism through which this process proceeds. A suggestion was made to the current authors (Tennyson, private communication, 1993) to investigate the effect of oxygen addition to the source gas on our measurements of H_3^+ recombination²¹. This ion is believed to have a stable triplet excited state, $^3\Sigma_u^+$, although this is in a linear configuration as opposed to the equilateral geometry of the ground state²². This experiment was performed but it was found that the addition of oxygen to the ion source produced no change in the measured cross section. This suggests, therefore, that the observed large recombination rate for H_3^+ cannot be explained as arising from an excited ion species.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the financial support of the Canadian Natural Sciences and Engineering Research Council and the United States Air Force Office of Scientific Research. M.Chibisov would also like to thank the Center for Chemical Physics at the University of Western Ontario for the award of Visiting Research Fellowship. The collaboration of A. Canosa was made possible by a NATO Exchange Program grant.

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Table 1(a). Energies E_v of the vibrational levels of the $a^3\Sigma^+$ state of HeH^+ , (given in eV with respect to the $\text{He}^+(1s) + \text{H}(1s)$ dissociation limit).

	$^4\text{HeH}^+$		$^3\text{HeH}^+$	
v	Quasi-classical	Quantal	Quasi-classical	Quantal
0	-0.0823	-0.0823	-0.0815	-0.0815
1	-0.0451	-0.0451	-0.0435	-0.0436
2	-0.0209	-0.0208	-0.0194	-0.0194
3	-0.0075	-0.0075	-0.0065	-0.0065
N_{max}	5	5	5	5

	$^4\text{HeD}^+$		$^3\text{HeD}^+$	
v	Quasi-classical	Quantal	Quasi-classical	Quantal
0	-0.0873	-0.0873	-0.0864	-0.0864
1	-0.0562	-0.0562	-0.0540	-0.0540
2	-0.0330	-0.0330	-0.0305	-0.0305
3	-0.0172	-0.0172	-0.0150	-0.0150
4	-0.0077	-0.0076	-0.0057	-0.0065
N_{max}	7	7	7	7

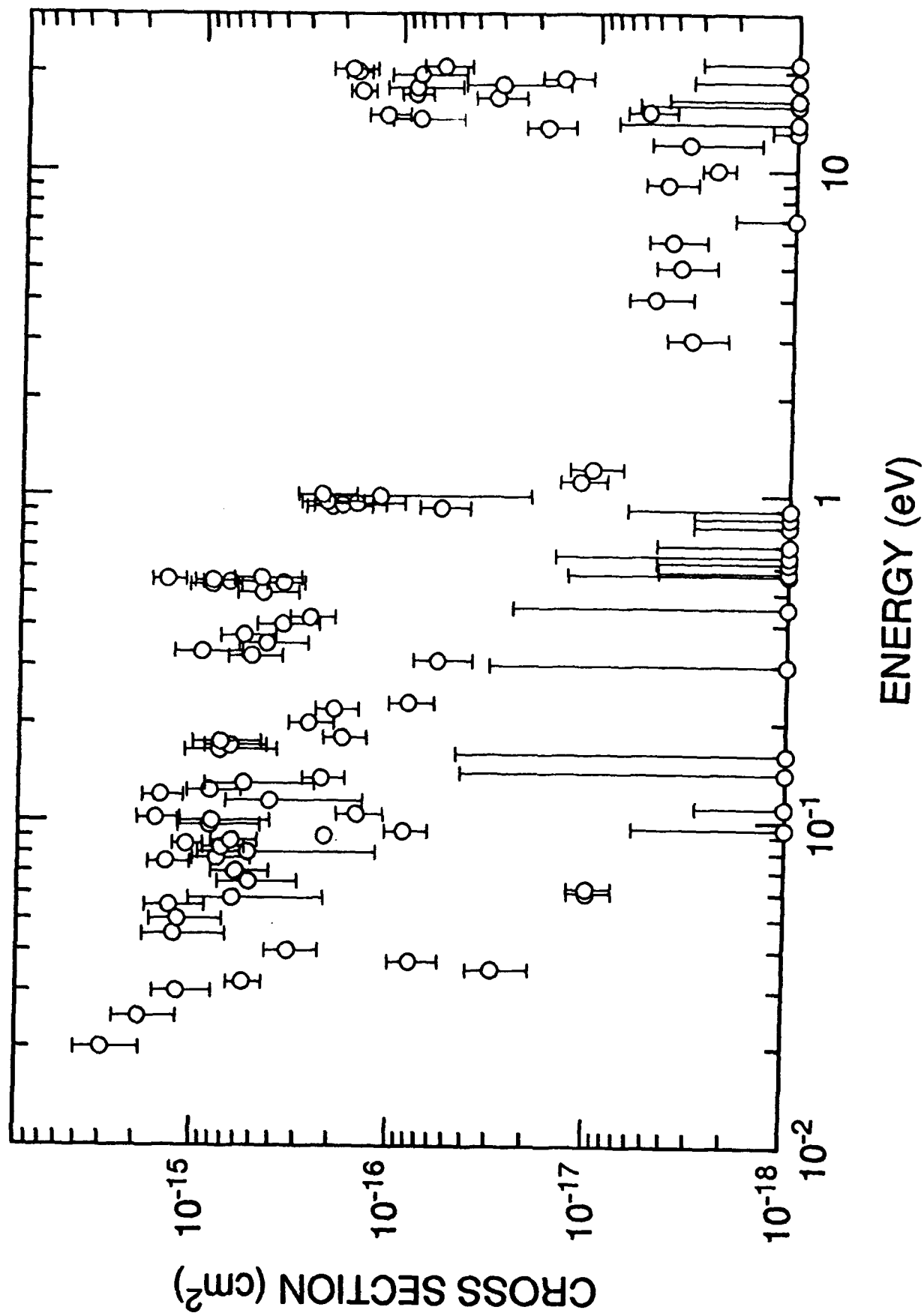
Table 1(b). Energies E_v of the vibrational levels of the $A^1\Sigma^+$ state of $^4\text{HeH}^+$.

v	0	1	2	3	
E_v	-0.0327	-0.0135	-0.0041	-0.0008	$N_{\text{max}}=4$

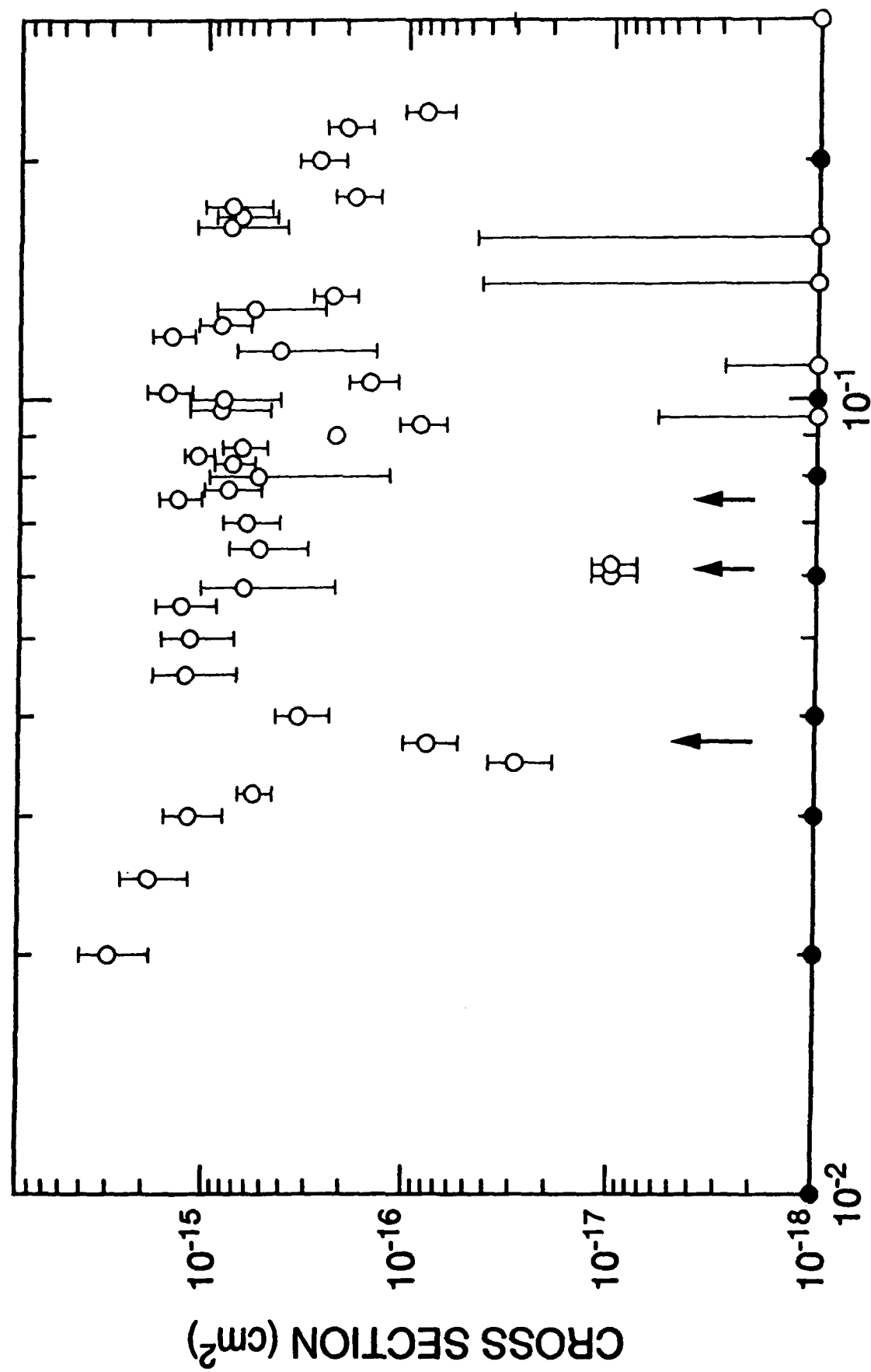
FIGURE CAPTIONS

1. Measured effective cross sections for the dissociative recombination of HeH^+ using a 50% H_2 /50% He source gas.
2. The low energy portion of figure 1 shown on an expanded scale. The energy separations between the excited and ground vibrational levels of the $a^3\Sigma^+$ state are shown by the arrows. The solid circles are measurements taken with a 25% H_2 :25%He:50% O_2 source gas.
3. The shallow potential wells of the $a^3\Sigma^+$ and the $A^1\Sigma^+$ states and their vibrational levels.
4. Schematic diagram of the curve crossings suitable for recombination of the $\text{HeH}^+ a^3\Sigma^+$ state. The insertion shows the reason for the recombination cross section going to zero above 0.55 eV.
5. The high energy portion of figure 1 shown on an expanded scale. The source gas was 50% H_2 /50% He.
6. High energy results obtained when the 50:50 H_2 :He source gas was replaced with a 25:25:50 H_2 :He: O_2 mixture.

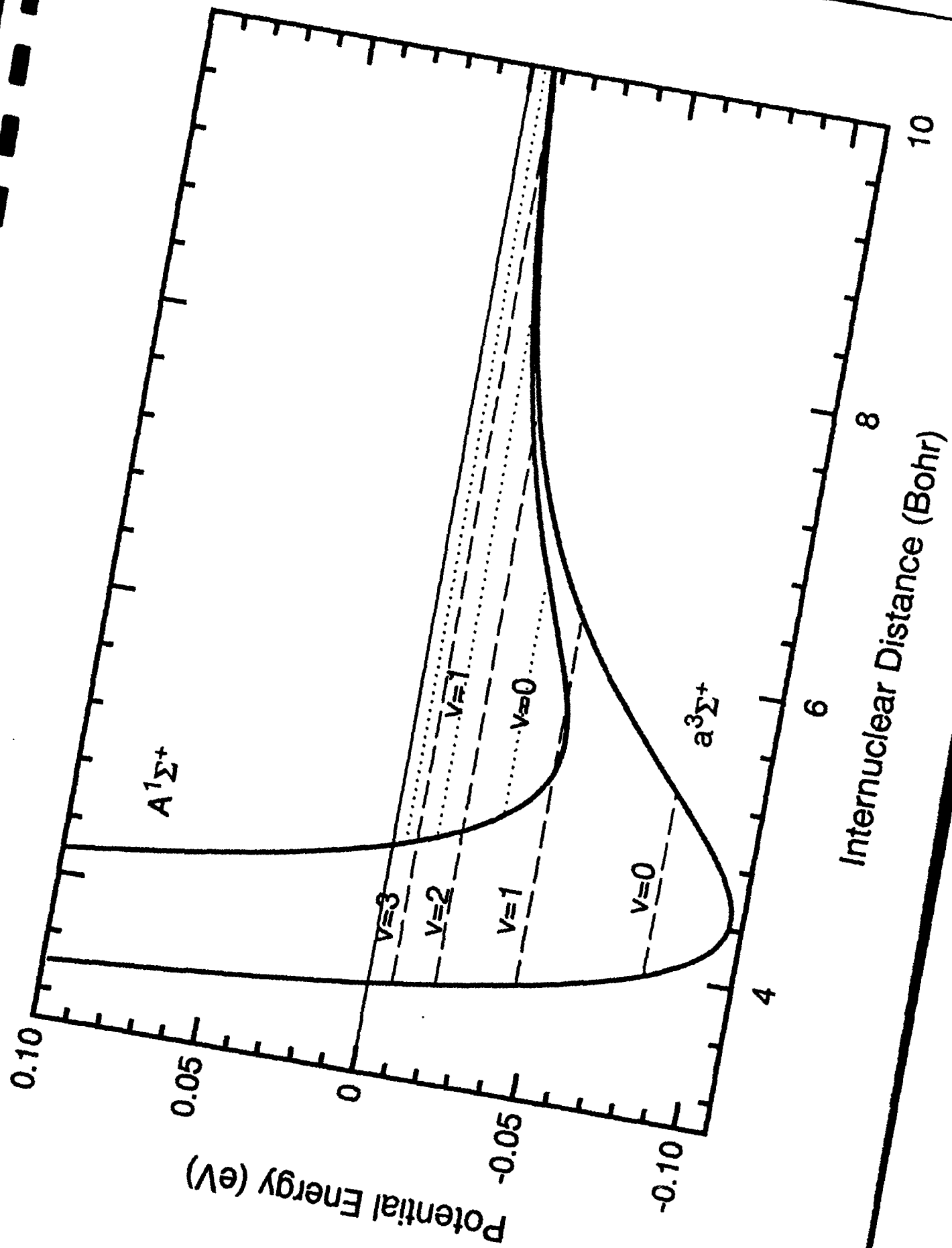
DR(HEH⁺)

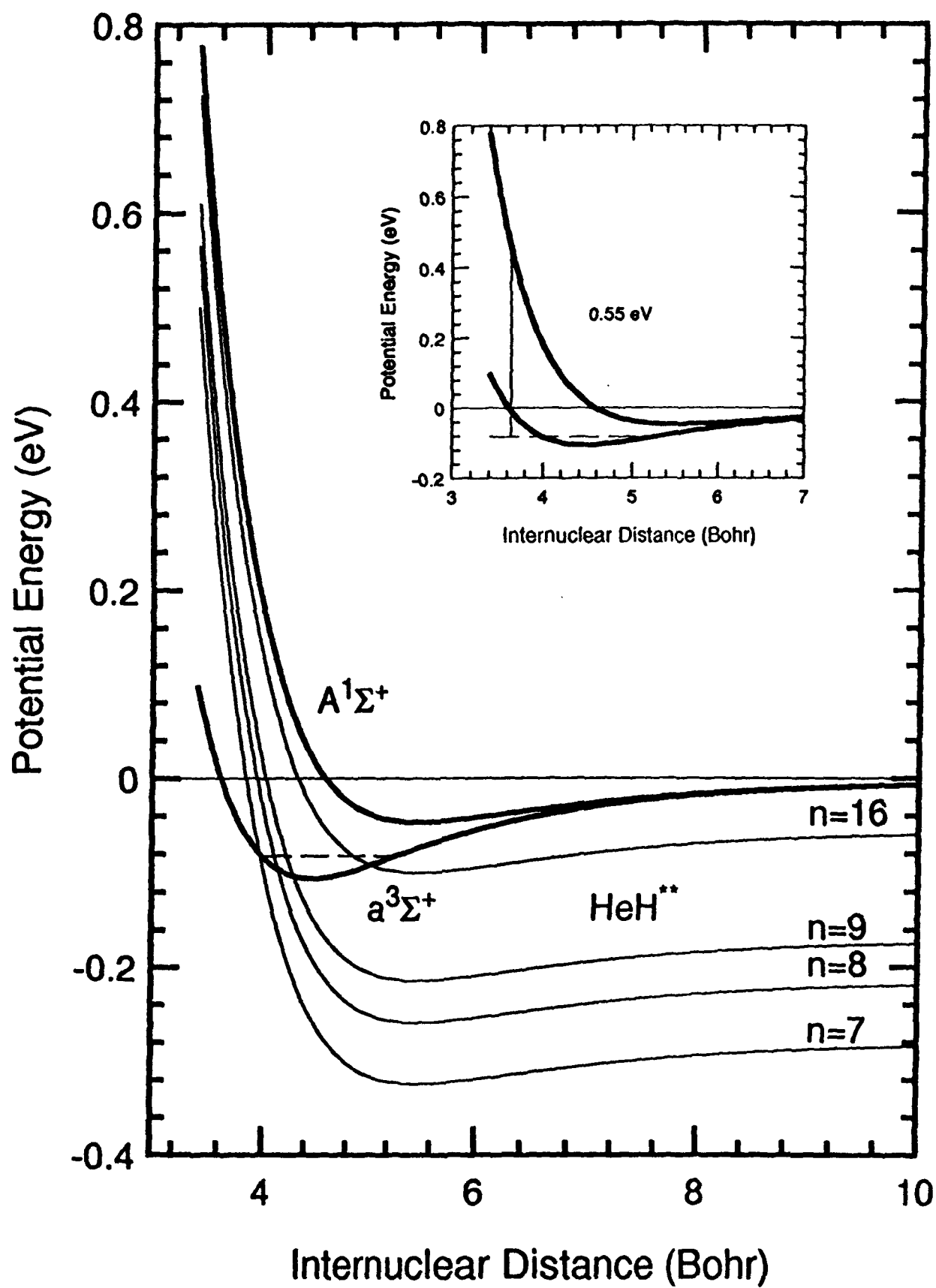


DR(HEH⁺)

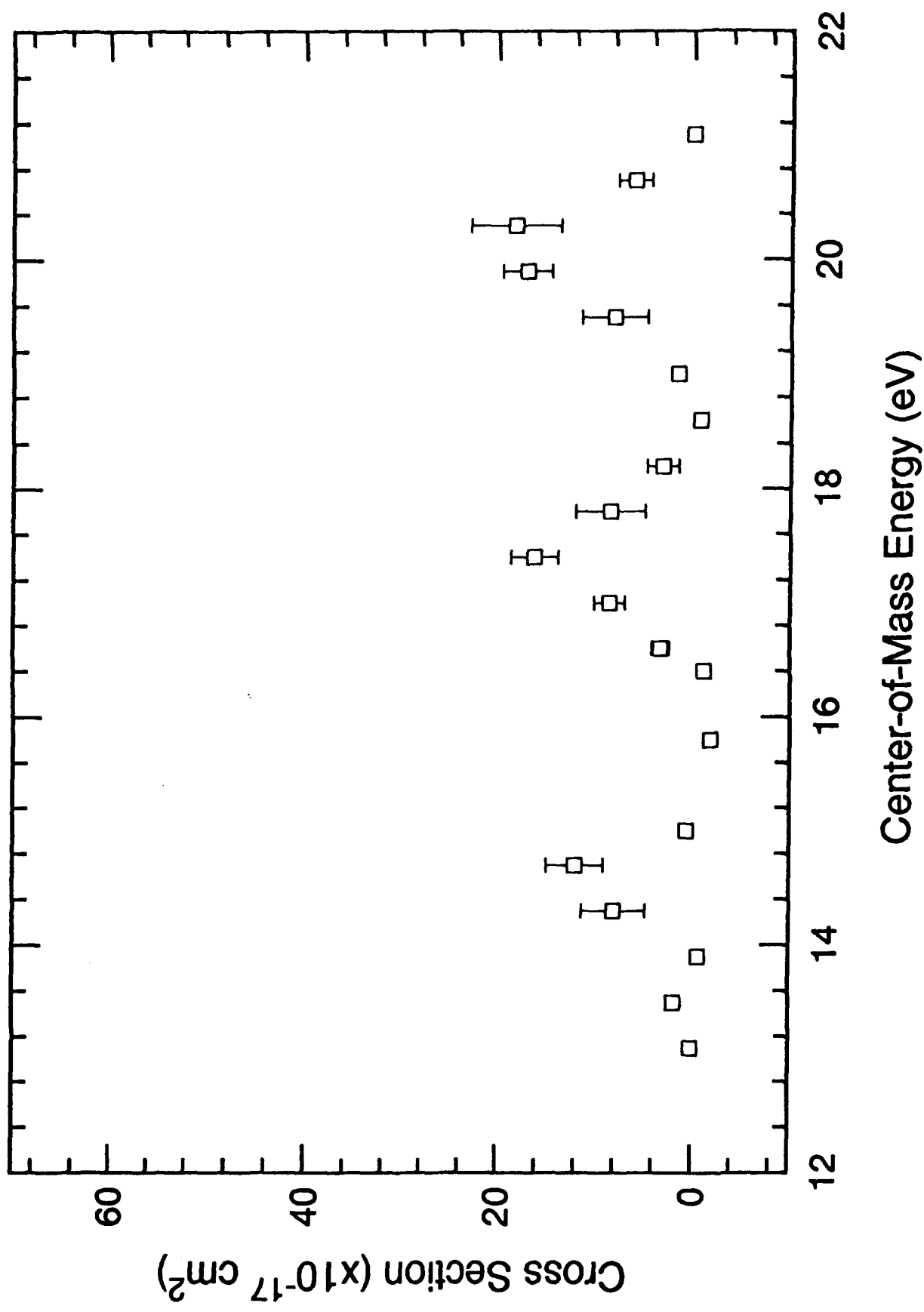


ENERGY (eV)





HEH⁺ Excited States



HEH⁺ GROUND STATE

